

Dynamics of quantum dissipation systems interacting with Fermion and Boson grand canonical bath ensembles: Hierarchical equations of motion approach

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A hierarchical equations of motion formalism for a quantum dissipation system in a grand canonical bath ensemble surrounding is constructed, on the basis of the calculus-on-path-integral algorithm, together with the parametrization of arbitrary non-Markovian bath that satisfies fluctuation-dissipation theorem. The influence functionals for both the Fermion or Boson bath interaction are found to be of the same path-integral expression as the canonical bath, assuming they all satisfy the Gaussian statistics. However, the equation of motion formalism are different, due to the fluctuation-dissipation theories that are distinct and used explicitly. The implications of the present work to quantum transport through molecular wires and electron transfer in complex molecular systems are discussed.

I. INTRODUCTION

Quantum dissipation theory (QDT) is concerned with the fundamental formulations for the dynamics of a quantum system of primary interest embedded in a quantum bath surrounding environment. The key quantity in QDT is the so-called reduced system density operator, $\rho(t) \equiv \text{tr}_B \rho_T(t)$, i.e., the partial trace of the total density operator over the bath space of practically infinite degrees of freedom. Due to its fundamental importance and intrinsic complexity, the development of QDT has involved scientists from many fields of research,^{1,2,3,4,5,6,7,8,9,10} but it has remained as a challenging topic since the middle of the last century.

The exact $\rho(t)$ can be expressed in terms of the Feynman-Vernon influence functional, assuming the interaction bath satisfies the Gaussian statistics.¹¹ Its numerical implementation has been carried out in a few simple systems with the forward-backward iterative path integral propagation method.^{12,13} Unraveling the path integral formalism into stochastic Schrödinger or stochastic Liouville equations has also been proposed in various forms in the past 10 years.^{14,15,16,17,18,19,20,21,22,23,24,25} The main obstacle to both the path integral and stochastic differential equation QDTs is their formidable numerical implementation in multilevel systems. Applications based on the path integral or stochastic QDT formalism are also often too cumbersome to be practical.

For the numerical efficiency and practical use in general, one will be interested in a differential equations of motion (EOM) formalism. In principle, a formally exact (but unclosed) EOM formalism of QDT can be constructed via the Nakajima-Zwanzig-Mori projection operator technique.^{26,27,28,29} To complete the formalism, however, one usually has to invoke certain approximations. In particular, various commonly used forms of Bloch-Redfield theory and Fokker-Planck equations^{30,31,32,33,34,35,36} involve the Markovian limit

and the weak system-bath interaction. The validities of these approximations are increasingly challenged, especially due to the emerging fields of nanoscience such as quantum transport and quantum information processes.^{37,38,39,40,41,42,43,44,45,46,47,48,49}

We have recently constructed a formally exact EOM formalism for arbitrary non-Markovian dissipation systems interacting with Gaussian canonical bath ensemble.^{50,51} This theory generalizes the Tanimura and coworkers' hierarchical formalism.^{52,53,54,55} The present paper is to extend our previous results to include both the Fermion and Boson grand canonical bath ensemble cases. The desired hierarchical EOM will be constructed, via the auxiliary influence-generating functionals (IGF) approach, together with the calculus-on-path-integrals (COPI) algorithm.^{50,51}

The remainder of this paper is organized as follows. In Sec. II, after the general description of a system-bath composite Hamiltonians, we discuss the fluctuation-dissipation theorem and other useful quantum mechanics relations for the grand canonical ensembles of Fermion/Boson particles. In Sec. III, we revisit the influence functionals path-integral formalism of QDT, assuming that the interaction grand canonical bath satisfies Gaussian statistics. The derivations in relation to these two sections are detailed in Appendix A and B, respectively. In Sec. IV, we present a bath correlation function parametrization scheme that will be used for the later development of differential EOM formalism for general non-Markovian dissipation systems. In Sec. V, we exploit a model dissipation system to illustrate the key ingredients in the IGF-COPI construction of the hierarchical EOM, augmented with residue correction and truncation. The final exact EOM formalism for general cases is presented in Sec. VI. It is in principle applicable for arbitrary non-Markovian dissipation systems, interacting with the Fermionic/Bosonic grand canonical bath ensemble at any temperature, including $T = 0$. Fi-

nally, Sec. VII presents the comments, discussions, and concluding remarks of this work.

II. TOTAL COMPOSITE HAMILTONIAN AND FLUCTUATION-DISSIPATION THEORY

A. The description of system–bath coupling

Let us start with the total system–plus–bath composite Hamiltonian, which in the stochastic bath h_B -interaction picture is given by

$$H_T(t) = H(t) + \sum_a \left[W_a \hat{f}_a^\dagger(t) + \hat{f}_a(t) W_a^\dagger \right]. \quad (1)$$

The deterministic Hamiltonian $H(t)$ for the reduced system may also involve a time-dependent external coherent field drive. The second term in the right-hand-side (rhs) of Eq. (1) denotes the stochastic system-bath interaction. It is expressed in the multiple dissipative mode decomposition form, where $\{W_a\}$ and $\{f_a(t) = e^{ih_B t} f_a e^{-ih_B t}\}$ are the system and bath operators, respectively. They are in general non-Hermitian.

The interaction bath operators $\{f_a(t)\}$ are also assumed to the Gaussian stochastic processes (*a la* the central limit theorem in statistics). The required Gaussian statistics is satisfied strictly when the individual f_a (f_a^\dagger) is a linear combination of the annihilation (creation) operators of the uncorrelated Fermion/Boson bath particles. The nonperturbative QDT formalisms to be developed later in this work will involve no further approximation.

Throughout this work, we set $\hbar \equiv 1$ and the inverse temperature $\beta \equiv 1/(k_B T)$. Denote $\langle \hat{O} \rangle_B \equiv \text{tr}_B(\hat{O} \rho_B^{\text{eq}})$, with the grand canonical equilibrium density operator of the bare bath being given by

$$\rho_B^{\text{eq}} = \frac{e^{-\beta(h_B - \mu \hat{N})}}{\text{tr}_B[e^{-\beta(h_B - \mu \hat{N})}]}. \quad (2)$$

Here, μ denotes the chemical potential of the bath. The particle number operator $\hat{N} = \sum_k c_k^\dagger c_k$ of the bath reservoirs commutes with the bare bath Hamiltonian, $[\hat{N}, h_B] = 0$. In the case of a canonical bath ensemble, the number of particles is conserved and no longer a dynamical variable. The resulting canonical $\rho_B^{\text{eq}} \propto e^{-\beta h_B}$ is equivalent to set the bath chemical potential $\mu = 0$.

For the later development, we also recast the system-bath coupling, i.e. the second term in the rhs of Eq. (1), as

$$H'(t) = \sum_{a,\sigma} W_a^\sigma \hat{f}_a^\sigma(t). \quad (3)$$

Here, $W_a^\sigma \equiv (W_a^\sigma)^\dagger$ that can be either W_a or W_a^\dagger ; and similarly for $\hat{f}_a^\sigma \equiv (\hat{f}_a^\sigma)^\dagger$. To be more specific, the bath operators $\{\hat{f}_a\}$ considered in this work are linear combinations of the annihilation operators, $\hat{f}_a = \sum_k t_{ak} c_k$,

with $c_j c_k^\dagger \pm c_k^\dagger c_j = \delta_{jk}$ for the Fermion (+) or Boson (−) particles in the grand canonical bath reservoirs.

Note that as the stochastic bath operators $\hat{f}_a^\sigma(t) = [\hat{f}_a^\sigma(t)]^\dagger$ are the linear combinations of either the creation or the annihilation operators of the particles, we have $\langle \hat{f}_a^\sigma(t) \rangle_B = \langle \hat{f}_a^\sigma(t) \hat{f}_b^\sigma(\tau) \rangle_B = 0$. These stochastic bath interaction operators are also assumed to follow the Wick's theorem for the thermodynamic Gaussian average that depends only on the two-time correlation functions,

$$C_{ab}^{(+)}(t - \tau) = \langle \hat{f}_a^\dagger(t) \hat{f}_b(\tau) \rangle_B, \quad (4a)$$

$$C_{ab}^{(-)}(t - \tau) = \langle \hat{f}_a(t) \hat{f}_b^\dagger(\tau) \rangle_B. \quad (4b)$$

B. Fermion versus Boson bath reservoirs: Fluctuation-dissipation theorem

Now we present the fluctuation-dissipation theorem (FDT), and the related symmetry and detailed-balance relations for the involving interaction bath correlation functions. The key steps to the final results will be detailed in Appendix A. To avoid the confusion of later using \pm to represent Fermion/Boson bath ensemble, let us recast Eqs. (4) as

$$C_{ab}^{(\sigma)}(t) = \langle \hat{f}_a^\sigma(t) \hat{f}_b^{\bar{\sigma}}(0) \rangle_B. \quad (5)$$

They satisfy the symmetry and the detailed-balance relations as [cf. Eqs. (A1)–(A7)]

$$[C_{ab}^{(\sigma)}(t)]^* = C_{ba}^{(\sigma)}(-t) = e^{\beta \sigma \mu} C_{ab}^{(\bar{\sigma})}(t - i\beta). \quad (6)$$

Their frequency-domain counterparts are

$$C_{ab}^{(\sigma)}(\omega) = [C_{ba}^{(\sigma)}(\omega)]^* = e^{\beta(\omega + \sigma \mu)} C_{ba}^{(\bar{\sigma})}(-\omega), \quad (7)$$

with the corresponding spectral functions of

$$C_{ab}^{(\sigma)}(\omega) \equiv \frac{1}{2} \int_{-\infty}^{\infty} dt e^{i\omega t} C_{ab}^{(\sigma)}(t). \quad (8)$$

The spectrum positivity that reads

$$C_{aa}^{(\sigma)}(\omega) \geq 0, \quad C_{aa}^{(\sigma)}(\omega) C_{bb}^{(\sigma)}(\omega) \geq |C_{ab}^{(\sigma)}(\omega)|^2, \quad (9)$$

can be readily verified via the same method used in the Appendix A of Ref. 36. The symmetry and detailed-balance relations [Eqs. (6) or Eqs. (7)] are general for a grand ensemble, no matter it is of Fermion or Boson in nature. This nature enters in terms of FDT via the spectral density functions as follows.

Consider $h_B = \sum_k \epsilon_k c_k^\dagger c_k$ and $\hat{f}_a = \sum_k t_{ak} c_k$. In this case, the interaction spectral density functions are defined physically for $\omega \geq \min(\epsilon_k) \equiv 0$ as

$$J_{ab}(\omega \geq 0) = \pi \sum_k t_{ak} t_{bk}^* \delta(\omega - \epsilon_k) = J_{ba}^*(\omega). \quad (10)$$

Mathematically, one can extend their definition to the $\omega < 0$ region, by setting $J_{ab}(\omega) \equiv \pm J_{ba}(-\omega)$ for the

Fermion/Boson (+/−) particles for the reasons that will become self-evident soon. As a result,

$$J_{ab}(\omega) = \pi \sum_k [t_{ak} t_{bk}^* \delta(\omega - \epsilon_k) \pm t_{ak}^* t_{bk} \delta(\omega + \epsilon_k)]. \quad (11)$$

On the other hand,

$$[\hat{f}_a(t), \hat{f}_b^\dagger(0)]_\pm = \sum_k t_{ak} t_{bk}^* e^{-i\epsilon_k t}, \quad (12a)$$

$$[\hat{f}_a^\dagger(t), \hat{f}_b(0)]_\pm = \pm \sum_k t_{ak}^* t_{bk} e^{i\epsilon_k t}, \quad (12b)$$

assuming c -numbers in this case. Consequently, Eq. (11) can be recast as

$$\begin{aligned} J_{ab}(\omega) &= \frac{1}{2} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [\hat{f}_a(t), \hat{f}_b^\dagger(0)]_\pm \rangle_B \\ &= \frac{1}{2} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [\hat{f}_a^\dagger(t), \hat{f}_b(0)]_\pm \rangle_B, \end{aligned} \quad (13)$$

or [cf. Eqs. (4) and (8)]

$$J_{ab}(\omega) = C_{ab}^{(\sigma)}(\omega) \pm C_{ba}^{(\bar{\sigma})}(-\omega). \quad (14)$$

Note the symmetry relations here,

$$J_{ba}(\omega) = \pm J_{ab}(-\omega) = J_{ab}^*(\omega). \quad (15)$$

From the second identity of Eq. (7), we have also

$$C_{ab}^{(\sigma)}(\omega) = \frac{J_{ab}(\omega)}{1 \pm e^{-\beta(\omega + \sigma\mu)}}, \quad (16)$$

or equivalently

$$C_{ab}^{(\sigma)}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t} J_{ab}(\omega)}{1 \pm e^{-\beta(\omega + \sigma\mu)}}. \quad (17)$$

This is the FDT for the Fermion (+) or Boson (−) grand canonical bath ensembles.

In concluding this section, let us make some comments on Eq. (14), which can in fact be considered as the working definition of the spectral density functions in general. For the linear bath interaction model, Eq. (14) is equivalent to the conventional expression, Eq. (10) or Eq. (11), in which $J_{ab}(\omega)$ is independent of temperature and chemical potential. As the working definition, Eq. (14) may also be useful even for the cases where the bath interaction depends nonlinearly on the annihilation (creation) operators of the Fermion/Boson bath ensembles. In such cases, Eq. (14) that does not depend on the σ -index may involve with a certain mean-field approximation and the resulting $J_{ab}(\omega)$ remains explicitly independent of temperature and chemical potential. The implicit dependence of $J_{ab}(\omega)$ on temperature and/or chemical potential may however arise from the mean-field values of t_{ak} or t_{bk} , as in Eq. (10) or (11).

Consider now the value of $J_{ab}(\omega = 0)$. As inferred from the second identity of Eq. (15), $J_{ab}(0) =$

$\pm J_{ab}^*(0)$, implying that $J_{ab}(0)$ is pure real/imaginary for a Fermion/Boson bath interaction. For the spectral density function, assuming it is analytical at $\omega = 0$, the Boson bath interaction will also assume

$$J_{ab}(0) = [(1 - e^{-\beta\omega}) C_{ab}^{(\sigma)}(\omega)]_{\omega \rightarrow 0; \mu=0} = 0; \quad (\text{Boson})$$

as inferred from Eq. (16) and from the fact that $J_{ab}(\omega)$ does not depend on μ explicitly.

We are interested in nonperturbative QDT, in the presence of non-Markovian multi-mode interaction with grand canonical Fermion/Boson bath. Both the path integral and the differential EOM formalisms of QDT will be considered. The former will be presented in the coming section, while the latter will be constructed in Sec. VI, on the basis of calculus (time-derivative) on the exact path integral expressions. In order to have the EOM hierarchically coupled and closed for a general non-Markovian case, the bath correlation functions $C_{ab}^{(\sigma)}(t)$ should be parameterized in a proper form. The aforementioned properties of $J_{ab}(\omega = 0)$ will be used there, together with Eqs. (15) and Eq. (17), to the FDT-preserved parametrization of $C_{ab}^{(\sigma)}(t)$ in Sec. IV that participates in the construction of the hierarchical EOM for general non-Markovian dissipation dynamics.

III. PATH INTEGRAL FORMALISM AND DISSIPATION FUNCTIONALS

In this section, we construct the path integral formalism of quantum dissipation. For the sake of clarity, we present only the final results here. The derivation of the path integral formalism will be given in Appendix B. It is similar to our previous work on the Bosonic canonical bath interaction cases, where the system and bath operators, $\{W_a\}$ and $\{f_a\}$, were also assumed to be Hermitian.^{50,51} Briefly speaking, it employs the Wick's theorem for the thermodynamic Gaussian average, followed by using the symmetry and detailed-balance relations of Eq. (6). It involves also the ansatz of the initially factorizable total density operator, $\rho_T(t_0) = \rho(t_0) \rho_B^{\text{eq}}$, with ρ_B^{eq} being given by Eq. (2). Note that when the initial time $t_0 \rightarrow -\infty$ this ansatz imposes no approximation.^{3,36}

Unlike its EOM counterpart that can be expressed in the operator level, the path integral formalism goes with a representation. Let $\{|\alpha\rangle\}$ be a basis set in the system subspace, and $\alpha \equiv (\alpha, \alpha')$ for abbreviation, so that $\rho(\alpha, t) \equiv \rho(\alpha, \alpha', t)$. Denote $\mathcal{U}(t, t_0)$ as the reduced Liouville-space propagator, $\rho(t) \equiv \mathcal{U}(t, t_0) \rho(t_0)$, which in the α -representation reads

$$\rho(\alpha, t) = \int d\alpha_0 \mathcal{U}(\alpha, t; \alpha_0, t_0) \rho(\alpha_0, t_0), \quad (18)$$

with the path integral expression of¹¹

$$\mathcal{U}(\alpha, t; \alpha_0, t_0) = \int_{\alpha_0[t_0]}^{\alpha[t]} \mathcal{D}\alpha e^{iS[\alpha]} \mathcal{F}[\alpha] e^{-iS[\alpha']}. \quad (19)$$

$S[\alpha]$ is the classical action functional of the reduced system, evaluated along a path $\alpha(\tau)$, with the constraints that two ending points $\alpha(t_0) = \alpha_0$ and $\alpha(t) = \alpha$ are fixed. The key quantity in the path integral formalism is the Feynman–Vernon influence functional $\mathcal{F}[\alpha]$ that will be presented soon.

In connection to the later development of EOM formalism, we denote hereafter $\mathbf{a} = (aa')$ for a pair of dissipation modes and introduce

$$\tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha\}) \equiv \tilde{W}_{aa'}^{(\sigma)}(t; \{\alpha\}) - \tilde{W}_{aa'}^{(\sigma)}(t; \{\alpha'\}), \quad (20)$$

where [noting that $\tilde{W}_{\mathbf{a}}^{(\sigma)} \equiv \tilde{W}_{aa'}^{(\sigma)}$ and $C_{\mathbf{a}}^{(\sigma)} \equiv C_{aa'}^{(\sigma)}$]

$$\tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha\}) \equiv \int_{t_0}^t d\tau C_{\mathbf{a}}^{(\sigma)}(t - \tau) W_{a'}^{\sigma}[\alpha(\tau)], \quad (21a)$$

$$\tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha'\}) \equiv \int_{t_0}^t d\tau [C_{\mathbf{a}}^{(\bar{\sigma})}(t - \tau)]^* W_{a'}^{\sigma}[\alpha'(\tau)]. \quad (21b)$$

Denote also

$$\mathcal{W}_{\mathbf{a}}^{\sigma}[\alpha] \equiv W_a^{\sigma}[\alpha(t)] - W_a^{\sigma}[\alpha'(t)] \equiv W_a^{\sigma} - W_a'^{\sigma}. \quad (22)$$

It is in fact the $W_{\mathbf{a}}^{(\sigma)}$ -commutator in the path-integral representation, as it depends only at the fixed ending points of the path. The time-dependence here can thus be removed.

The final expressions for the influence functional are [cf. Eq. (B7)]

$$\mathcal{F}[\alpha] \equiv \exp \left\{ - \int_{t_0}^t d\tau \mathcal{R}[\tau; \{\alpha\}] \right\}, \quad (23a)$$

with

$$\mathcal{R}[t; \{\alpha\}] \equiv \sum_{\mathbf{a}, \sigma} \mathcal{W}_{\mathbf{a}}^{\bar{\sigma}}[\alpha] \tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha\}). \quad (23b)$$

Here, \mathcal{F} is expressed in terms of its exponent time-integrand \mathcal{R} . It is in fact the *time-local dissipation super-operator* in the path integral representation, as the time derivative of Eq. (23a),

$$\partial_t \mathcal{F} = -\mathcal{R} \mathcal{F}, \quad (24)$$

leads to $\partial_t \mathcal{U} = -i\mathcal{L}\mathcal{U} - \mathcal{R}(t)\mathcal{U}$ [cf. Eq. (19)], or equivalently $\dot{\rho} = -i\mathcal{L}\rho - \mathcal{R}(t)\rho$ [cf. Eq. (18)]. Therefore, \mathcal{R} of Eq. (23b) can be termed as the *dissipation functional*.

It is noticed that the chemical potential μ does not appear explicitly in the dissipation functional \mathcal{R} . The above exact path integral formalism, Eqs. (18)–(23), is formally the same for both the canonical and the grand canonical bath interactions, no matter the bath is of Fermion or Boson, as long as the Gaussian bath statistics is applicable. Apparently, in the case of $W_a = W_a^{\dagger} \equiv Q_a$ and $\hat{f}_a = \hat{f}_a^{\dagger} = \hat{F}_a$, the sign-index σ (“+” and “−”) can be omitted, and Eqs. (20)–(23) recovers formally the Eqs. (10)–(13) of Ref. 51, where the Bosonic canonical bath ensembles were also assumed. The different nature of the quantum bath ensemble is embedded implicitly in the correlation functions $C_{\mathbf{a}}^{(\sigma)}(t)$, via the FDT and symmetry relations as described in Sec. II B; see Eqs. (15) and (17).

IV. NON-MARKOVIAN BATH VIA PARAMETERIZATION

To construct a hierarchically coupled set of EOM for a general non-Markovian dissipation, $C_{\mathbf{a}}^{(\sigma)}(t)$ should be expressed in a proper form, such as an exponential series expansion,^{49,50,51,56} which shall also satisfy the FDT [Eq. (17)]. To that end, let us consider the following parametrization form on the bath spectral density functions ($J_{\mathbf{a}} \equiv J_{aa'}$),

$$J_{\mathbf{a}}(\omega) = J_{\mathbf{D}}^{\mathbf{a}}(\omega) + \sum_{k=1}^K \left[\frac{\zeta_k^{\mathbf{a}} \gamma_k^{\mathbf{a}} + i \bar{\zeta}_k^{\mathbf{a}} \omega}{(\omega - \omega_k^{\mathbf{a}})^2 + (\gamma_k^{\mathbf{a}})^2} \pm \frac{\zeta_k^{\mathbf{a}} \gamma_k^{\mathbf{a}} + i \bar{\zeta}_k^{\mathbf{a}} \omega}{(\omega + \omega_k^{\mathbf{a}})^2 + (\gamma_k^{\mathbf{a}})^2} \right], \quad (25a)$$

with the Drude term

$$J_{\mathbf{D}}^{\mathbf{a}}(\omega) = \frac{\zeta_{\mathbf{D}}^{\mathbf{a}} \gamma_{\mathbf{D}}^{\mathbf{a}} + i \bar{\zeta}_{\mathbf{D}}^{\mathbf{a}} \omega}{\omega^2 + (\gamma_{\mathbf{D}}^{\mathbf{a}})^2} \quad \text{or} \quad \frac{\zeta_{\mathbf{D}}^{\mathbf{a}} \omega}{\omega^2 + (\gamma_{\mathbf{D}}^{\mathbf{a}})^2}. \quad (25b)$$

for the Fermion (+) or Boson (−) bath ensembles, respectively.

All involving parameters are real; $\omega_k^{\mathbf{a}}$ and $\gamma_k^{\mathbf{a}}$ (including $\gamma_{\mathbf{D}}^{\mathbf{a}}$) are positive as well. They satisfy [cf. Eq. (15)] $(\omega_k^{aa'}, \gamma_k^{aa'}, \zeta_k^{aa'}, \bar{\zeta}_k^{aa'}) = (\omega_k^{a'a}, \gamma_k^{a'a}, \zeta_k^{a'a}, -\bar{\zeta}_k^{a'a})$. The corresponding bath correlation functions can then be obtained via the FDT of Eq. (17), using the contour integration method. The final results read ($M \rightarrow \infty$)

$$C_{\mathbf{a}}^{(\sigma)}(t) = \eta_{\mathbf{D}}^{(\mathbf{a})} e^{-\gamma_{\mathbf{D}}^{\mathbf{a}} t} + \sum_{j=1}^{2K} \eta_j^{(\mathbf{a})} \phi_j^{\mathbf{a}}(t) + \sum_{m=1}^M \check{C}_m^{(\mathbf{a})}(t). \quad (26)$$

Hereafter, $\eta_j^{(\mathbf{a})} \equiv \eta_{\mathbf{a},j}^{(\sigma)}$ and $\eta_j^{(\bar{\mathbf{a}})} \equiv \eta_{\mathbf{a},j}^{(\bar{\sigma})}$ and similarly for other parameters that depend both (σ) and \mathbf{a} .

The first two terms in the rhs of Eq. (26), where

$$\phi_{2k-1}^{\mathbf{a}}(t) \equiv \sin(\omega_k^{\mathbf{a}} t) \exp(-\gamma_k^{\mathbf{a}} t), \quad (27a)$$

$$\phi_{2k}^{\mathbf{a}}(t) \equiv \cos(\omega_k^{\mathbf{a}} t) \exp(-\gamma_k^{\mathbf{a}} t), \quad (27b)$$

arise from the poles of $J_{\mathbf{a}}(z)$ in Eq. (25). The involving η -coefficients are

$$\eta_{\mathbf{D}}^{(\mathbf{a})} = \frac{\zeta_{\mathbf{D}}^{\mathbf{a}} + \bar{\zeta}_{\mathbf{D}}^{\mathbf{a}}}{e^{i\beta(\gamma_{\mathbf{D}}^{\mathbf{a}} + \sigma i\mu)} + 1} \quad \text{or} \quad \frac{i\zeta_{\mathbf{D}}^{\mathbf{a}}}{e^{i\beta(\gamma_{\mathbf{D}}^{\mathbf{a}} + \sigma i\mu)} - 1},$$

for the Fermion (+) or Boson (−), respectively; $\eta_{2k-1}^{(\mathbf{a})} = -i\lambda_k^{\mathbf{a}} \nu_k^{(\mathbf{a})} \pm i\bar{\lambda}_k^{\mathbf{a}} \nu_k^{(\mathbf{a})}$ and $\eta_{2k}^{(\mathbf{a})} = \lambda_k^{\mathbf{a}} \bar{\nu}_k^{(\mathbf{a})} \pm \bar{\lambda}_k^{\mathbf{a}} \nu_k^{(\mathbf{a})}$. Here, $\lambda_k^{\mathbf{a}} \equiv [(\zeta_k^{\mathbf{a}} + \bar{\zeta}_k^{\mathbf{a}}) \gamma_k^{\mathbf{a}} + \bar{\zeta}_k^{\mathbf{a}} \omega_k^{\mathbf{a}}] / (2\gamma_k^{\mathbf{a}}) \equiv (\bar{\lambda}_k^{\mathbf{a}})^*$, while $\nu_k^{(\mathbf{a})} \equiv \{1 \pm \exp[\beta(\omega_k^{\mathbf{a}} + i\gamma_k^{\mathbf{a}} - \sigma\mu)]\}^{-1}$, and $\bar{\nu}_k^{(\mathbf{a})}$ is similar as $\nu_k^{(\mathbf{a})}$ but with $-\omega_k^{\mathbf{a}}$ instead.

The last term of Eq. (26) arises from the poles of Matsubara frequencies,

$$\tilde{\gamma}_m \equiv (2m - 1)\pi/\beta \quad \text{or} \quad 2m\pi/\beta \quad (28)$$

for the Fermion or Boson bath ensemble, respectively.

$$\check{C}_m^{(a)}(t) \equiv \check{\eta}_m^{(a)} \exp[-(\check{\gamma}_m - \sigma i\mu)t] = \mp[\check{C}_m^{(\bar{a})}(t)]^*, \quad (29)$$

with

$$\check{\eta}_m^{(a)} \equiv -i(2/\beta)J_a(-i\check{\gamma}_m - \sigma\mu) = \mp[\check{\eta}_m^{(\bar{a})}]^*. \quad (30)$$

The last identity in each of the above two equations is originated from $J_a(-z) = \pm[J_a(z)]^*$, the analytical continuation of $J_a(-\omega) = \pm J_a^*(\omega)$ [cf. Eqs.(15)]. It will show that this property leads to some simplification in treating the Matsubara-frequency contributions. In the canonical ensemble, the Matsubara coefficients $\check{\eta}_m^{(a)}$ of Eq. (30), which do not depend on σ and chemical potential μ , are real and pure imaginary for Boson and Fermion bath ensembles, respectively.

To conclude this section, let us make some comments on the parameterized $C_a^{(\sigma)}(t)$ of Eq. (26), for which the hierarchical EOM formalism can be constructed via the IGF-COPI method^{50,51} without approximations (cf. Sec. VI). In principle, Eq. (26) can be exact for a general non-Markovian dissipation, if the involving K and M are sufficiently large. The resulting EOM formalism is also exact; but its size grows in a power law as K or M increases. The exact evaluation of complex dissipation would rapidly become extremely tedious. In practice, the K and M in Eq.(26) have to be finite in the construction of the EOM formalism. To complete the theory, the residue correction due to the difference between the true and the parameterized correlation functions, $\delta C_a^{(\sigma)} \equiv C_{a,\text{exa}}^{(\sigma)} - C_a^{(\sigma)}$, should also be incorporated into the final EOM formalism.

V. APPROACH TO THE EXACT EQUATIONS OF MOTION FORMALISM: PRINCIPLES

A. Hierarchy construction

For the sake of clarity, we exploit the broadband dissipation case to illustrate the IGF-COPI approach to the hierarchical EOM, together with the residue correction and hierarchy truncation that will be demonstrated in the next subsection. In the broadband limit, $\eta_b^{(a)} \exp(-\gamma_b^{(a)}t) \rightarrow \eta_b^{(a)} \gamma_b^{(a)} \delta(t)$ and the $\eta_j^{(a)}$ -term in Eq. (26) can be completely neglected. We will show in Sec. VB that the $\delta(t)$ -like component can be easily taken into account in terms of the residue correction.

Considered here explicitly for the illustration of hierarchy construction are only the Matsubara terms,

$$C_a^{(\sigma)}(t) = \sum_{m=1}^M \check{C}_m^{(a)}(t) = \sum_{m=1}^M \check{\eta}_m^{(a)} e^{-(\check{\gamma}_m - \sigma i\mu)t}. \quad (31)$$

The dissipative functional \mathcal{R} [Eq. (23b)], as it appears linearly in the bath correlation functions, can be expanded

like Eq. (31) as

$$\mathcal{R} = \sum_{a,\sigma,m} \mathcal{W}_a^{\bar{\sigma}}[\alpha] \check{W}_m^{(a)}(t; \{\alpha\}). \quad (32)$$

Here, $\check{W}_m^{(a)}$ are defined by Eqs. (20) and (21) but with the bath correlation functions being replaced by $\check{C}_m^{(a)}$. We shall hereafter omit the explicit path-integral variables dependence whenever it does not cause confusion.

Note that the Matsubara terms possess two special properties: $\check{C}_m^{(a)}(t) = \pm[\check{C}_m^{(\bar{a})}(t)]^*$ [cf. Eq. (29)] and the fact that their time constants $(\check{\gamma}_m - \sigma i\mu)$ are dissipation-mode independent. As a result, Eq. (32) can have the summation over a' performed, resulting in

$$\mathcal{R} = i \sum_{a,\sigma,m} \mathcal{W}_a^{\bar{\sigma}} \check{Z}_m^{(a)}. \quad (33)$$

The involving Eqs. (20) and (21) are rearranged for the Matsubara terms as

$$\check{Z}_m^{(a)} \equiv -i \sum_{a'} \check{W}_m^{(a)} = -i \left(\check{W}_m^{(a)} \pm \check{W}_m'^{(a)} \right), \quad (34)$$

with

$$\check{W}_m^{(a)} \equiv \sum_{a'} \int_{t_0}^t d\tau \check{C}_m^{(a)}(t-\tau) W_{a'}^{\sigma}[\alpha(\tau)], \quad (35a)$$

$$\check{W}_m'^{(a)} \equiv \sum_{a'} \int_{t_0}^t d\tau \check{C}_m^{(a)}(t-\tau) W_{a'}^{\sigma}[\alpha'(\tau)]. \quad (35b)$$

We have

$$\partial_t \check{Z}_m^{(a)} = \check{C}_m^{(a)} - (\check{\gamma}_m - \sigma i\mu) \check{Z}_m^{(a)}, \quad (36)$$

where

$$\check{C}_m^{(a)} \equiv -i \sum_{a'} \check{\eta}_m^{(a)} (W_{a'}^{\sigma} \pm W_{a'}'^{\sigma}). \quad (37)$$

We are now in the position to construct the EOM via the calculus (time-derivative) on the path integral formalism.^{50,51} The time derivative on the action functional parts contributes to the coherent dynamics of $-i\mathcal{L}U$, and thus can be included into the final EOM formalism. However, the time derivative of the influence functional, Eq.(24), will generate other *auxiliary influence functionals* (AIFs). This process goes on progressively and hierarchically as the time derivatives on AIFs is continued. It is also noticed that time derivative of individual $\check{Z}_m^{(a)}$ is closed by itself [Eq. (36)]. We will see soon that this property, together with their exclusive relation to the dissipation functional as Eq.(33), make $\{\check{Z}_m^{(a)}\}$ the complete set of the IGFs for the desired hierarchy construction.

Let us start with the time derivative on the primary influence functional, Eq. (24). In the present demonstrative case, it reads

$$\partial_t \mathcal{F} = -\mathcal{R}\mathcal{F} = -i \sum_{a,\sigma,m} \mathcal{W}_a^{\bar{\sigma}} \mathcal{F}_m^{(a)}. \quad (38)$$

Generated here are the first-tier AIFs, defined as

$$\mathcal{F}_m^{(a)} \equiv \check{Z}_m^{(a)} \mathcal{F}. \quad (39)$$

Compared with the primary \mathcal{F} [Eq. (23)] whose leading term is 1, the first-tier AIFs $\{\mathcal{F}_m^{(a)}\}$ are of the second-order in the system–bath coupling as their leading contributions.

To proceed, the time derivatives on those first-tier AIFs are also needed. We obtain [cf. Eqs. (36)–(39)],

$$\partial_t \mathcal{F}_m^{(a)} = \left[\check{\mathcal{C}}_m^{(a)} \mathcal{F} - (\hat{\gamma}_m - \sigma i \mu) \mathcal{F}_m^{(a)} \right] + \check{Z}_m^{(a)} (\partial_t \mathcal{F}). \quad (40)$$

Here,

$$\check{Z}_m^{(a)} (\partial_t \mathcal{F}) = -i \sum_{a', \sigma', m'} \mathcal{W}_{a'}^{\sigma'} \mathcal{F}_{mm'}^{(aa')}, \quad (41)$$

with (denoting $\check{Z}_{m'}^{(a')} \equiv \check{Z}_{a', m'}^{(\sigma')}$)

$$\mathcal{F}_{mm'}^{(aa')} \equiv \check{Z}_m^{(a)} \mathcal{F}_{m'}^{(a')} = (\check{Z}_m^{(a)} \check{Z}_{m'}^{(a')}) \mathcal{F}, \quad (42)$$

being the next tier (second-tier) AIFs. Apparently, the leading contributions in $\mathcal{F}_{mm'}^{(aa')}$ are of the fourth-order in the system–bath coupling.

Continue on applying Eqs. (36)–(38) for Eq. (42)

$$\partial_t \mathcal{F}_{mm'}^{(aa')} = [\partial_t (\check{Z}_m^{(a)} \check{Z}_{m'}^{(a')})] \mathcal{F} + (\check{Z}_m^{(a)} \check{Z}_{m'}^{(a')}) (\partial_t \mathcal{F}). \quad (43)$$

The first term in the rhs above will lead to the dependence on the tier-minus-one AIFs, arising from the $\check{\mathcal{C}}_m^{(a)}$ –term of Eq. (36), while the $(\partial_t \mathcal{F})$ –term above leads to the tier-plus-one AIFs.

Apparently, those distinct $\{\check{Z}_m^{(a)}\}$ are IGFs, since all involving AIFs can be generically expressed as

$$\mathcal{F}_{mm' \dots m''}^{(aa' \dots a'')} = (\check{Z}_m^{(a)} \check{Z}_{m'}^{(a')} \dots \check{Z}_{m''}^{(a'')}) \mathcal{F}. \quad (44)$$

In order to eliminate possible duplications due to the indexes permutation/repetition that result in the same AIF, we arrange all distinct $\{\check{Z}_m^{(a)}\}$ in a specified sequence and rearrange Eq. (44) accordingly as

$$\mathcal{F}_n \equiv \left\{ \prod_{a, \sigma, m} (\check{Z}_m^{(a)})^{\check{n}_m^{(a)}} \right\} \mathcal{F}. \quad (45)$$

Here $\mathbf{n} = \{\check{n}_m^{(a)}\}$ is the index-set of nonnegative integers for the number of occurrences on the distinct $\check{Z}_m^{(a)}$ individuals in the specified sequence. Note that the total number of nonnegative integers in the index-set \mathbf{n} is $2Mq$, where M is the number of Matsubara terms considered, q the number of dissipative mode a , while the factor 2 arises from $\sigma = +, -$.

To specify the hierarchical dependence of $\partial_t \mathcal{F}_n$, we denote also the index-set $\check{n}_{(a), m}^{\pm} \equiv \{\dots, \check{n}_m^{(a)} \pm 1, \dots\}$ that deviates from \mathbf{n} only by changing the specified $\check{n}_m^{(a)}$ to

$\check{n}_m^{(a)} \pm 1$. The time derivative of \mathcal{F}_n [Eq. (45)] can be carried out, again by using Eqs. (36) and (38), resulting in

$$\begin{aligned} \partial_t \mathcal{F}_n = & -(\check{\gamma}_n + i \check{n} \mu) \mathcal{F}_n + \sum_{a, \sigma, m} \check{n}_m^{(a)} \check{\mathcal{C}}_m^{(a)} \mathcal{F}_{\check{n}_{(a)}^-} \\ & -i \sum_{a, \sigma, m} \mathcal{W}_a^{\bar{\sigma}} \mathcal{F}_{\check{n}_{(a), m}^+}. \end{aligned} \quad (46)$$

Here,

$$\check{\gamma}_n \equiv \sum_{a, \sigma, m} \check{n}_m^{(a)} \check{\gamma}_m = \sum_{a, m} [\check{n}_{a, m}^{(-)} + \check{n}_{a, m}^{(+)}] \check{\gamma}_m, \quad (47a)$$

$$\check{n} \equiv - \sum_{a, \sigma, m} \sigma \check{n}_m^{(a)} = \sum_{a, m} [\check{n}_{a, m}^{(-)} - \check{n}_{a, m}^{(+)}]. \quad (47b)$$

The auxiliary reduced density operators can now be defined as (including also $\rho \equiv \rho_0$)

$$\rho_n(t) \equiv \mathcal{U}_n(t, t_0) \rho(t_0), \quad (48)$$

with the auxiliary propagators of [cf. Eq. (19)]

$$\mathcal{U}_n(\alpha, t; \alpha_0, t_0) \equiv \int_{\alpha_0}^{\alpha} \mathcal{D}\alpha e^{iS[\alpha]} \mathcal{F}_n[\alpha] e^{-iS[\alpha']}. \quad (49)$$

The hierarchically coupled Eqs. (46) can now be recast as

$$\begin{aligned} \dot{\rho}_n = & -[i(\mathcal{L} + \check{n} \mu) + \check{\gamma}_n] \rho_n + \sum_{a, \sigma, m} \check{n}_m^{(a)} \check{\mathcal{C}}_m^{(a)} \rho_{\check{n}_{(a)}^-} \\ & -i \sum_{a, \sigma, m} \mathcal{W}_a^{\bar{\sigma}} \rho_{\check{n}_{(a), m}^+}. \end{aligned} \quad (50)$$

Here, $\mathcal{W}_a^{\bar{\sigma}}$ and $\check{\mathcal{C}}_m^{(a)}$, which were given in the path integral representation as Eqs. (22) and (37), respectively, are now the reduced Liouville-space operators; i.e.,

$$\mathcal{W}_a^{\sigma} \hat{O} = [\mathcal{W}_a^{\sigma}, \hat{O}], \quad \check{\mathcal{C}}_m^{(a)} \hat{O} = -i \sum_{a'} \check{\gamma}_m^{(a)} [\mathcal{W}_{a'}^{\sigma}, \hat{O}]_{\pm}. \quad (51)$$

B. Residue correction and the hierarchy truncation

We have illustrated the IGF-COPI approach to the hierarchical EOM formalism. The same method will be used in Sec. VI to establish the EOM formalism for a general non-Markovian dissipation characterized by Eq. (26) without approximation. The final hierarchical EOM [cf. Eq. (74)] can be expressed as

$$\dot{\rho}_n = -[i(\mathcal{L} + \check{n} \mu) + \gamma_n + \delta \mathcal{R}(t)] \rho_n + \rho_n^{\{\equiv\}} + \rho_n^{\{-\}} + \rho_n^{\{+\}}. \quad (52)$$

The hierarchy-down $\rho_n^{\{-\}}$ and hierarchy-up $\rho_n^{\{+\}}$ are similar as the last two terms in Eq. (50), but with the Drude and the ϕ_j^{α} –components of the bath correlation functions of Eq. (26) included. The hierarchical-swap $\rho_n^{\{\equiv\}}$ arises completely from the ϕ_j^{α} –components, thus have no correspondence in Eq. (50).

Involved in Eq. (52) is also the residue correction $\delta\mathcal{R}(t)$ to the dissipation. It accounts for the practical difference between the exact $C_{\mathbf{a},\text{exa}}^{(\sigma)}(t)$ and the parameterized $C_{\mathbf{a}}^{(\sigma)}(t)$ by Eq. (26) where K and M are finite. In the following we shall present the principle of residue correction⁵¹ to construct the residue dissipation $\delta\mathcal{R}$, followed by an efficient scheme of hierarchy truncation.

Let us first verify that the residue dissipation correction $\delta\mathcal{R} = \mathcal{R}_{\text{exa}} - \mathcal{R}$, due to $\delta C_{\mathbf{a}}^{(\sigma)} \equiv C_{\mathbf{a},\text{exa}}^{(\sigma)} - C_{\mathbf{a}}^{(\sigma)}$, does enter at the global level, participating in every tier of the hierarchy as Eq. (52). As inferred from Eq. (23a), the exact influence functional of primary interest reads in path integral representation as $\mathcal{F}^{\text{exa}} = \mathcal{F}\mathcal{F}_{\text{resi}}$, with $\partial_t\mathcal{F} = -\mathcal{R}\mathcal{F}$ and $\partial_t\mathcal{F}_{\text{resi}} = -\delta\mathcal{R}\mathcal{F}_{\text{resi}}$. The auxiliary influence functionals defined, for example in Eq. (45), for the construction of the hierarchical EOM should now be replaced by $\mathcal{F}_n^{\text{exa}} \equiv \mathcal{F}_n\mathcal{F}_{\text{resi}}$. Its time derivative is then $\partial_t\mathcal{F}_n^{\text{exa}} = (\partial_t\mathcal{F}_n)\mathcal{F}_{\text{resi}} - \delta\mathcal{R}\mathcal{F}_n^{\text{exa}}$. Together with the established $\partial_t\mathcal{F}_n$ such as Eq. (46) before residue correction, the above expression verifies the fact that $\delta\mathcal{R}$ does enter into every tier of the final hierarchical EOM, as in Eq. (52).

We are now in the position to present some practically useful expressions of $\delta\mathcal{R}(t)$. Note that its exact expression in path integral representation is [cf. Eq. (23b) with Eqs. (20) and (21)]

$$\delta\mathcal{R} = \sum_{\mathbf{a},\sigma} \mathcal{W}_{\mathbf{a}}^{\bar{\sigma}}[\alpha] \delta\tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha\}), \quad (53)$$

where

$$\delta\tilde{W}_{\mathbf{a}}^{(\sigma)} = \delta\tilde{W}_{\mathbf{a}}^{(\sigma)}(t; \{\alpha\}) - \delta\tilde{W}_{\mathbf{a}}^{\prime(\sigma)}(t; \{\alpha'\}), \quad (54)$$

with

$$\delta\tilde{W}_{\mathbf{a}}^{(\sigma)} = \int_{t_0}^t d\tau \delta C_{\mathbf{a}}^{(\sigma)}(t - \tau) W_{\mathbf{a}'}^{\sigma}[\alpha(\tau)], \quad (55a)$$

$$\delta\tilde{W}_{\mathbf{a}}^{\prime(\sigma)} = \int_{t_0}^t d\tau [\delta C_{\mathbf{a}}^{(\bar{\sigma})}(t - \tau)]^* W_{\mathbf{a}'}^{\sigma}[\alpha'(\tau)]. \quad (55b)$$

In the Markovian-residue limit, Eqs. (55) reduce to

$$\delta\tilde{W}_{\mathbf{a}}^{(\sigma)} \approx \delta\bar{C}_{\mathbf{a}}^{(\sigma)}(t) W_{\mathbf{a}'}^{\sigma}[\alpha(t)], \quad (56)$$

and similar for $\delta\tilde{W}_{\mathbf{a}}^{\prime(\sigma)}$; with

$$\delta\bar{C}_{\mathbf{a}}^{(\sigma)}(t) = \int_{t_0}^t d\tau \delta C_{\mathbf{a}}^{(\sigma)}(t - \tau). \quad (57)$$

Note that the system variable $W_{\mathbf{a}'}^{\sigma}$ in Eqs. (56) is now evaluated at the ending time of the path integral where $\alpha(t) = \alpha$ and $\alpha'(t) = \alpha'$ are fixed. As results, Eq. (53) for the Markovian-residue dissipation can be expressed in the operator level as

$$\delta\mathcal{R}\hat{O} = \sum_{\mathbf{a},\sigma} [W_{\mathbf{a}}^{\bar{\sigma}}, \delta\bar{C}_{\mathbf{a}}^{(\sigma)}(t) W_{\mathbf{a}'}^{\sigma} \hat{O} - [\delta\bar{C}_{\mathbf{a}}^{(\bar{\sigma})}(t)]^* \hat{O} W_{\mathbf{a}'}^{\sigma}]. \quad (58)$$

Alternatively, one can exploit various second-order QDT formalism³⁶ for the residue $\delta\mathcal{R}$ via, for example,

$$\delta\tilde{W}_{\mathbf{a}}^{(\sigma)}(t) \approx \int_{t_0}^t d\tau \delta C_{\mathbf{a}}^{(\sigma)}(t - \tau) e^{-i\mathcal{L}(t-\tau)} W_{\mathbf{a}'}^{\sigma}. \quad (59)$$

The dissipation-free propagator (assuming also time-independent system Hamiltonian for clarity) is used here to connect $W_{\mathbf{a}'}^{\sigma}[\alpha(\tau)]$ in Eq. (55) to its value at the fixed ending path point of $\alpha(t) = \alpha$. It is the reason that the α -representation has been removed from Eq. (59), as it is now treated as an operator; the same for $\delta\tilde{W}_{\mathbf{a}}^{\prime(\sigma)}$. Therefore, the weak-residue $\delta\mathcal{R}$ assumes the operator form as

$$\delta\mathcal{R}\hat{O} = \sum_{\mathbf{a},\sigma} [W_{\mathbf{a}}^{\bar{\sigma}}, \delta\tilde{W}_{\mathbf{a}}^{(\sigma)}(t)\hat{O} - \hat{O}\delta\tilde{W}_{\mathbf{a}}^{\prime(\sigma)}(t)]. \quad (60)$$

Apparently, the Markovian-residue limit is the special case of the weak-residue treatment.

To the end of this section, let us show that the principle of residue correction presented above can also be used for the hierarchy truncation. Recall the hierarchical AIF considered in the previous subsection, \mathcal{F}_n of Eq. (45). Implied there is a trivial identity that

$$\mathcal{F}_{\mathbf{n}(a),m}^{+} = \check{Z}_m^{(a)} \mathcal{F}_n = -i(\check{W}_m^{(a)} \pm \check{W}_m^{\prime(a)}) \mathcal{F}_n. \quad (61)$$

Note that $\check{W}_m^{(a)}$ and $\check{W}_m^{\prime(a)}$ defined in Eqs. (35) as their path integral expressions are of the same mathematical structure as Eq. (55). Therefore, the afore-described method, via either the Markovian or second-order approximation scheme to the explicit operator form of $\check{W}_m^{(a)}$, can be adopted *locally* at the desired anchoring level \mathbf{N} . Thus, the truncation scheme of

$$\rho_{\mathbf{N}(a),m}^{+} = \check{Z}_m^{(a)} \rho_{\mathbf{N}} = -i[\check{W}_m^{(a)}, \rho_{\mathbf{N}}]_{\pm}, \quad (62)$$

in which $\check{W}_m^{(a)}$ assumes its approximate operator form is established. The choice of anchoring index-set \mathbf{N} will be specified at the concluding part of the coming section, where the hierarchical EOM formalism for the general non-Markovian dissipation is treated.

VI. EQUATIONS OF MOTION THEORY FOR GENERAL NON-MARKOVIAN DISSIPATION

A. Influence generating functionals

We are now in the position to the IGF-COPI construction of the exact hierarchical EOM formalism for the parameterized $C_{\mathbf{a}}^{(\sigma)}$ of Eq. (26). The corresponding dissipation functional can be expressed as [cf. Eqs. (23b) and (33)]

$$\begin{aligned} \mathcal{R} = & i \sum_{\mathbf{a},\sigma} \mathcal{W}_{\mathbf{a}}^{\bar{\sigma}} Z_{\mathbf{D}}^{(a)} + i \sum_{\mathbf{a},\sigma,k} \mathcal{W}_{\mathbf{a}}^{\bar{\sigma}} [X_k^{(a)} + Y_k^{(a)}] \\ & + i \sum_{\mathbf{a},\sigma,m} \mathcal{W}_{\mathbf{a}}^{\bar{\sigma}} \check{Z}_m^{(a)}. \end{aligned} \quad (63)$$

The last term arising from the Matsubara contribution had been treated in detail in Sec. V A, see Eqs. (33)–(37). Apparently, all these composite $\{X, Y, Z\}$ -functionals are IGFs; but whether they are completed or not should be checked with their time derivatives; see Sec. V A.

Consider the Drude-IGFs in Eq. (63), they are given by

$$Z_D^{(\mathbf{a})} \equiv -i \left(\eta_D^{(\mathbf{a})} \tilde{W}_D^{(\mathbf{a})} - \eta_D^{(\bar{\mathbf{a}})*} \tilde{W}_D'^{(\mathbf{a})} \right), \quad (64a)$$

with

$$\tilde{W}_D^{(\mathbf{a})} = \int_{t_0}^t d\tau e^{-\gamma_D^{(\mathbf{a})}(t-\tau)} W_{a'}^\sigma[\alpha(\tau)]. \quad (64b)$$

We obtain

$$\partial_t Z_D^{(\mathbf{a})} = \mathcal{C}_D^{(\mathbf{a})} - \gamma_D^{(\mathbf{a})} Z_D^{(\mathbf{a})}. \quad (65)$$

Here

$$\mathcal{C}_D^{(\mathbf{a})} \equiv -i \left(\eta_D^{(\mathbf{a})} W_{a'}^\sigma - \eta_D^{(\bar{\mathbf{a}})*} W_{a'}'^\sigma \right). \quad (66)$$

Therefore, $\{Z_D^{(\mathbf{a})}\}$ constitute the complete IGFs as the Drude-term contribution is concerned.

Turn now to the $\{X, Y\}$ -IGFs. They are given by

$$X_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k}^{(\mathbf{a})} \tilde{W}_{2k}^{(\mathbf{a})} - \eta_{2k}^{(\bar{\mathbf{a}})*} \tilde{W}_{2k}'^{(\mathbf{a})} \right), \quad (67a)$$

$$Y_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k-1}^{(\mathbf{a})} \tilde{W}_{2k-1}^{(\mathbf{a})} - \eta_{2k-1}^{(\bar{\mathbf{a}})*} \tilde{W}_{2k-1}'^{(\mathbf{a})} \right), \quad (67b)$$

with [see Eq. (27) for $\phi(t)$]

$$\tilde{W}_j^{(\mathbf{a})} = \int_{t_0}^t d\tau \phi_j^{(\mathbf{a})}(t-\tau) W_{a'}^\sigma[\alpha(\tau)]. \quad (68)$$

Unlike the Z -functionals for the Drude and Matsubara components, the time derivatives of the X - and Y -functionals are closed, together with two additional functionals, given by

$$\bar{X}_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k}^{(\mathbf{a})} \tilde{W}_{2k-1}^{(\mathbf{a})} - \eta_{2k}^{(\bar{\mathbf{a}})*} \tilde{W}_{2k-1}'^{(\mathbf{a})} \right), \quad (69a)$$

$$\bar{Y}_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k-1}^{(\mathbf{a})} \tilde{W}_{2k}^{(\mathbf{a})} - \eta_{2k-1}^{(\bar{\mathbf{a}})*} \tilde{W}_{2k}'^{(\mathbf{a})} \right). \quad (69b)$$

We have

$$\partial_t X_k^{(\mathbf{a})} = \mathcal{A}_k^{(\mathbf{a})} - \gamma_k^{(\mathbf{a})} X_k^{(\mathbf{a})} - \omega_k^{(\mathbf{a})} \bar{X}_k^{(\mathbf{a})}, \quad (70a)$$

$$\partial_t \bar{X}_k^{(\mathbf{a})} = \omega_k^{(\mathbf{a})} X_k^{(\mathbf{a})} - \gamma_k^{(\mathbf{a})} \bar{X}_k^{(\mathbf{a})}; \quad (70b)$$

and

$$\partial_t \bar{Y}_k^{(\mathbf{a})} = \mathcal{B}_k^{(\mathbf{a})} - \gamma_k^{(\mathbf{a})} \bar{Y}_k^{(\mathbf{a})} - \omega_k^{(\mathbf{a})} Y_k^{(\mathbf{a})}, \quad (70c)$$

$$\partial_t Y_k^{(\mathbf{a})} = \omega_k^{(\mathbf{a})} \bar{Y}_k^{(\mathbf{a})} - \gamma_k^{(\mathbf{a})} Y_k^{(\mathbf{a})}. \quad (70d)$$

Here

$$\mathcal{A}_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k}^{(\mathbf{a})} W_{a'}^\sigma - \eta_{2k}^{(\bar{\mathbf{a}})*} W_{a'}'^\sigma \right), \quad (71a)$$

$$\mathcal{B}_k^{(\mathbf{a})} \equiv -i \left(\eta_{2k-1}^{(\mathbf{a})} W_{a'}^\sigma - \eta_{2k-1}^{(\bar{\mathbf{a}})*} W_{a'}'^\sigma \right). \quad (71b)$$

These six classes of (XYZ) -functionals constitute now a complete set of IGFs for the hierarchy construction. The general expression for all auxiliary influence functionals in the hierarchy can then be expressed as

$$\mathcal{F}_n = \left\{ \prod_{\mathbf{a},k} \left[\left(Y_k^{(\mathbf{a})} \right)^{n_{2k-1}^{(\mathbf{a})}} \left(X_k^{(\mathbf{a})} \right)^{n_{2k}^{(\mathbf{a})}} \left(\bar{Y}_k^{(\mathbf{a})} \right)^{\bar{n}_{2k-1}^{(\mathbf{a})}} \left(\bar{X}_k^{(\mathbf{a})} \right)^{\bar{n}_{2k}^{(\mathbf{a})}} \right] \right. \\ \left. \times \prod_{\mathbf{a},\sigma} \left(Z_D^{(\mathbf{a})} \right)^{n_D^{(\mathbf{a})}} \prod_{\mathbf{a},\sigma,m} \left(\tilde{Z}_m^{(\mathbf{a})} \right)^{\tilde{n}_m^{(\mathbf{a})}} \right\} \mathcal{F}. \quad (72)$$

The index in \mathcal{F}_n is specified by the involving nonnegative integers,

$$\mathbf{n} \equiv \left(n_j^{(\mathbf{a})}, \bar{n}_j^{(\mathbf{a})}, n_D^{(\mathbf{a})}, \tilde{n}_m^{(\mathbf{a})}; j = 1, \dots, 2K; m = 1, \dots, M \right). \quad (73)$$

Therefore, the total number of the nonnegative integers in the index-set \mathbf{n} is $2(p + 4Kp + Mq)$, where q denotes the number of dissipative modes, and $p \leq q^2$ the number of nonzero dissipative mode pairs. In terms of Eqs. (48) and (49), the IGF-COPI approach to the hierarchical EOM is now completed by considering the time derivative of \mathcal{F}_n [Eq. (72)], which can be readily carried out using Eqs. (24), (36), (65) and (70).

B. Hierarchical equations of motion

The final hierarchical EOM for the associating auxiliary density operators read

$$\dot{\rho}_n = -[i(\mathcal{L} + \tilde{n}\mu) + \gamma_n] \rho_n + \rho_n^{\{\equiv\}} + \rho_n^{\{-\}} + \rho_n^{\{+\}}. \quad (74)$$

The μ -term in Eq. (74) arises from the corresponding contribution of Matsubara term [Eq. (36)] given by Eq. (47b). The γ -terms in Eq. (74) arise from the damping-terms of Eqs. (70). The resulting damping constant is given by

$$\gamma_n \equiv \sum_{\mathbf{a},k} \left[n_{2k}^{(\mathbf{a})} + \bar{n}_{2k}^{(\mathbf{a})} + n_{2k-1}^{(\mathbf{a})} + \bar{n}_{2k-1}^{(\mathbf{a})} \right] \gamma_k^{(\mathbf{a})} \\ + \sum_{\mathbf{a},\sigma} n_D^{(\mathbf{a})} \gamma_D^{(\mathbf{a})} + \sum_{\mathbf{a},\sigma,m} \tilde{n}_m^{(\mathbf{a})} \tilde{\gamma}_m^{(\mathbf{a})}. \quad (75)$$

The second term in Eq. (74) stems from the (off-diagonal) swap-terms of Eqs. (70). It reads

$$\rho_n^{\{\equiv\}} = \sum_{\mathbf{a},\sigma,k} \omega_k^{(\mathbf{a})} \left(n_{2k-1}^{(\mathbf{a})} \rho_{n_{(\mathbf{a}),2k-1}}^{\rightarrow} - n_{2k}^{(\mathbf{a})} \rho_{n_{(\mathbf{a}),2k}}^{\rightarrow} \right. \\ \left. - \bar{n}_{2k-1}^{(\mathbf{a})} \rho_{n_{(\mathbf{a}),2k-1}}^{\leftarrow} + \bar{n}_{2k}^{(\mathbf{a})} \rho_{n_{(\mathbf{a}),2k}}^{\leftarrow} \right). \quad (76)$$

The index-set $n_{(\mathbf{a}),j}^{\rightarrow}$ and $n_{(\mathbf{a}),j}^{\leftarrow}$ differ from \mathbf{n} of Eq. (73) only at the specified $\{(\mathbf{a}), j\}$ by

$$n_{(\mathbf{a}),j}^{\rightarrow} : (n_j^{(\mathbf{a})}, \bar{n}_j^{(\mathbf{a})}) \rightarrow (n_j^{(\mathbf{a})} - 1, \bar{n}_j^{(\mathbf{a})} + 1),$$

$$n_{(\mathbf{a}),j}^{\leftarrow} : (n_j^{(\mathbf{a})} + 1, \bar{n}_j^{(\mathbf{a})} - 1) \leftarrow (n_j^{(\mathbf{a})}, \bar{n}_j^{(\mathbf{a})}).$$

The third term in Eq. (74) stems from the $(\mathcal{A}, \mathcal{B}, \mathcal{C})$ -terms of Eqs. (70), while the last term is from Eq. (24).

They are the hierarchy-down and hierarchy-up contributions, respectively, and given by

$$\rho_n^{\{-\}} = \sum_{\mathbf{a}, \sigma, k} \left[n_{2k}^{(\mathbf{a})} \mathcal{A}_k^{(\mathbf{a})} \rho_{n_{(\mathbf{a}), 2k}^-} + \bar{n}_{2k-1}^{(\mathbf{a})} \mathcal{B}_k^{(\mathbf{a})} \rho_{\bar{n}_{(\mathbf{a}), 2k-1}^-} \right] + \sum_{\mathbf{a}, \sigma} n_D^{(\mathbf{a})} \mathcal{C}_D^{(\mathbf{a})} \rho_{n_{(\mathbf{a}), D}^-} + \sum_{\mathbf{a}, \sigma, m} \check{n}_m^{(\mathbf{a})} \check{\mathcal{C}}_m^{(\mathbf{a})} \rho_{\check{n}_{(\mathbf{a}), m}^-}, \quad (77)$$

and ($j = D, 1, \dots, 2K$)

$$\rho_n^{\{+\}} \equiv -\mathcal{R}\rho_n = -i \sum_{\mathbf{a}, \sigma, j} \mathcal{W}_a^{\bar{\sigma}} \rho_{n_{(\mathbf{a}), j}^+} - i \sum_{\mathbf{a}, \sigma, m} \mathcal{W}_a^{\bar{\sigma}} \rho_{\bar{n}_{(\mathbf{a}), m}^+}. \quad (78)$$

The reduced Liouville-space operator \mathcal{W}_a^{σ} in Eq. (78) and $\check{\mathcal{C}}_m^{(\mathbf{a})}$ involved in Eq. (77) were given by Eq. (51). In Eq. (77), $\mathcal{A}_k^{(\mathbf{a})}$, $\mathcal{B}_k^{(\mathbf{a})}$, and $\mathcal{C}_D^{(\mathbf{a})}$ denote the reduced Liouville-space operator counterparts of Eqs. (66) and (71)

$$\mathcal{A}_k^{(\mathbf{a})} \hat{O} = -i(\eta_{2k}^{(\mathbf{a})} W_{a'}^{\sigma} \hat{O} - \eta_{2k}^{(\bar{\mathbf{a}})*} \hat{O} W_{a'}^{\sigma}), \quad (79a)$$

$$\mathcal{B}_k^{(\mathbf{a})} \hat{O} = -i(\eta_{2k-1}^{(\mathbf{a})} W_{a'}^{\sigma} \hat{O} - \eta_{2k-1}^{(\bar{\mathbf{a}})*} \hat{O} W_{a'}^{\sigma}), \quad (79b)$$

$$\mathcal{C}_D^{(\mathbf{a})} \hat{O} \equiv -i(\eta_D^{(\mathbf{a})} W_{a'}^{\sigma} \hat{O} - \eta_D^{(\bar{\mathbf{a}})*} \hat{O} W_{a'}^{\sigma}), \quad (79c)$$

The index-set $n_{(\mathbf{a}), j}^{\pm}$ ($\bar{n}_{(\mathbf{a}), j}^{\pm}$ or $\check{n}_{(\mathbf{a}), m}^{\pm}$) differs from \mathbf{n} only by changing the specified $n_j^{(\mathbf{a})}$ ($\bar{n}_j^{(\mathbf{a})}$ or $\check{n}_m^{(\mathbf{a})}$) to $n_j^{(\mathbf{a})} \pm 1$ ($\bar{n}_j^{(\mathbf{a})} - 1$ or $\check{n}_m^{(\mathbf{a})} \pm 1$). Note that the $(\bar{n}_{(\mathbf{a}), j}^+)^{\text{th}}$ -auxiliary reduced density operators are not generated from Eq. (78), since \bar{X}_k and \bar{Y}_k do not appear in the influence phase integrand \mathcal{R} [Eq. (24)]; they are rather generated from the EOM for the $(n_{(\mathbf{a}), j}^+)^{\text{th}}$ -auxiliary reduced density operators via the involved swap $\{\rightleftharpoons\}$ -terms [cf. Eq. (76)].

Note that, in Eq. (74), the reduced density matrix $\rho_0 = \rho$ is of primary interest, and the initial conditions are $\rho_n(t_0) = \rho(t_0)\delta_{n0}$, as inferred from their definitions. Setting the initial time $t_0 \rightarrow -\infty$, the established Hierarchical EOM is imposed no approximation. The initial conditions are now $\dot{\rho}(t_0) = 0$, where t_0 can be at any time before applying the the external time-dependent fields. The pulse-field induced dynamics will then be evaluated via Eq. (74) in the presence of external field.

C. Comments

1. Residue dissipation

The hierarchical EOM, Eqs. (74)–(79), are exact for a general dissipation system that involves the parameterized bath correlation functions of Eqs. (26). Due to the finite K and M of Eqs. (26) in practice, the complete theory can be established by considering the residue dissipation $\delta\mathcal{R}(t)$, due to the small difference between the exact and the parameterized $C_{\mathbf{a}}^{(\sigma)}(t)$ as described in section VB. The final hierarchical EOM thus read [cf. Eq. (52)]

$$\dot{\rho}_n = -[i(\mathcal{L} + \check{n}\mu) + \gamma_n + \delta\mathcal{R}(t)]\rho_n + \rho_n^{\{\rightleftharpoons\}} + \rho_n^{\{-\}} + \rho_n^{\{+\}}. \quad (80)$$

Actually, this hierarchical EOM Eq. (80) is valid for arbitrary temperature, including $T = 0$, at which the Matsubara damping constant $\check{\gamma}_m$ is meaningless and Matsubara expansion is not valid. Based on the principle of the residue correction, considering the finite temperature in the Matsubara term, the difference between the exact ($T = 0$) and the parameterized $C_{\mathbf{a}}^{(\sigma)}(t)$ (finite T) can be incorporated in the residue dissipation $\delta\mathcal{R}(t)$.

In the high temperature limit, a large Matsubara damping constant $\check{\gamma}_m \propto kT$ leads to the Markovian-limit bath correlation of $\check{C}_m^{\mathbf{a}}(t)$. In this case, the Matsubara-contribution can also be included in the residue dissipation $\delta\mathcal{R}(t)$ as given by Eq. (58) which enter the theory globally at every hierarchical tier.

2. Truncation consideration

The hierarchy truncation via the principle of residue correction has also been demonstrated in subsection VB, where only the Matsubara term is considered. In addition to the truncation of the Matsubara term of Eqs. (61) and (62), the same procedure is applied to the Drude and the ϕ terms ($\rho_{N_{(\mathbf{a}), D}^+}$ and $\rho_{N_{(\mathbf{a}), j}^+}$).

Now we consider the choice of the anchor index-set \mathbf{N} . The anchoring indexes can be specified for the individual constituents of the interaction bath correlation functions $C_{\mathbf{a}}^{(\sigma)}(t)$ of Eq. (26) as

$$N_D^{\mathbf{a}}, N_k^{\mathbf{a}}, \check{N}_m^{\mathbf{a}}; \quad \text{with } k = 1, \dots, K+1; \quad m = 1, \dots, M.$$

The closed set of hierarchically coupled EOM will then contain those ρ_n , whose individual index-set consists of the nonnegative integers that are confined within

$$\sum_{\sigma} n_{2k}^{(\mathbf{a})} + n_{2k+1}^{(\mathbf{a})} + \bar{n}_{2k}^{(\mathbf{a})} + \bar{n}_{2k+1}^{(\mathbf{a})} \leq N_k^{\mathbf{a}}, \quad (81a)$$

$$\sum_{\sigma} n_D^{(\mathbf{a})} \leq N_D^{\mathbf{a}}, \quad \sum_{\sigma} \check{n}_m^{(\mathbf{a})} \leq \check{N}_m^{\mathbf{a}}. \quad (81b)$$

The anchor index-set \mathbf{N} in $\rho_{\mathbf{N}}$ can now be defined as those with at least one upper limit being reached. The truncation can thus be made based on the formally exact relations such as Eqs. (61) and (62).

VII. CONCLUDING REMARKS

In summary, we have generalized the exact non-Markovian quantum dissipation theory,^{50,51} on the basis of the calculus-on-path-integral algorithm, to non-hermit coupling and non-Markovian grand canonical bath ensembles (both for Fermion and Boson statistics). The resulting dissipation functionals path-integral expression is found to be of the same as the canonical bath with the assumption of they all satisfying the Gaussian statistics. The distinct fluctuation dissipation properties lead to different hierarchical EOM coupling structures. However, the difference appears only in the Matsubara

contributions, Boson-commutator versus the Fermion-anticommutator relations, due to the particularity of the Matsubara coefficients [Eq. (30)]. In the high temperature limit, the aforementioned distinction diminishes, as both the Fermion and Boson bath ensembles approach to the Boltzmann statistics.

The hierarchical EOM formalism, which is in principle equivalent to its path-integral counterpart, may be relatively tractable; see the Summary of Ref. 51. Its numerical implementation remains a challenge, especially for complicated, strong dissipation systems at the extremely low temperature regime. Note that the hierarchy is not just for system-bath coupling strength, but more importantly also for bath correlation time scale.⁵⁰ The proper choice of system to effectively reduce the non-Markovian system-bath coupling strength in Eq. (1) is therefore still important both physically and numerically here. The standard approaches in this regard include the variation-principle based canonical transformation (e.g. the polaron picture) and the primary-bath-mode method (e.g. the solvation coordinate picture).^{57,58,59,60,61}

The present theory can account for the particle transfer between the system and the bath. Thus the problems of quantum transport through molecular wires and electron transfer in complex molecular systems can be demonstrated by this theory. For instance, if W_a (W_a^\dagger) represents annihilation (creation) operator of the system in the coupling Hamiltonian [Eq. (3)], the measurable quantity, such as the current can be exactly expressed as $\langle \hat{I}(t) \rangle = -i \langle [\hat{N}, H_T(t)] \rangle_T = -i \text{Tr}_S \sum_a \{ W_a \rho_a^\dagger(t) - W_a^\dagger \rho_a(t) \}$. Here $\rho_a(t)$ [$\rho_a^\dagger(t)$] is the summation over the reduced density matrixes in the first-tier described by Eq. (74) with $n = 1$. This detail work will be treated elsewhere. The present theory can be further generalized to the system coupled with many reservoirs (including phonon bath). The work along this line is in progress and will be published elsewhere.

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APPENDIX A: QUANTUM STATISTICAL PROPERTIES OF THE INTERACTION BATH CORRELATION FUNCTIONS: DERIVATION

The symmetry relation – the first identity of Eq. (6). This can be obtained immediately from the definition of correlation functions, Eq. (5), together with the time-translation invariance and the trace cyclic invariance.

The details are as follows [noting that $\hat{f}_a^\sigma \equiv (\hat{f}_a^\sigma)^\dagger$].

$$\begin{aligned} [C_{ab}^{(\sigma)}(t)]^* &\equiv \text{tr}_B[e^{ih_B t} \hat{f}_a^\sigma e^{-ih_B t} \hat{f}_b^\sigma \rho_B^{\text{eq}}]^* \\ &= \text{tr}_B[\rho_B^{\text{eq}} \hat{f}_b^\sigma e^{ih_B t} \hat{f}_a^\sigma e^{-ih_B t}] \\ &= \text{tr}_B[\hat{f}_b^\sigma(0) \hat{f}_a^\sigma(t) \rho_B^{\text{eq}}] \\ &= C_{ba}^{(\sigma)}(-t). \end{aligned} \quad (\text{A1})$$

The detailed-balance relation – the second identity of Eq. (6). Its derivation exploits the property of

$$e^{\beta\mu\hat{N}} \hat{f}_a^\sigma e^{-\beta\mu\hat{N}} = e^{\sigma\beta\mu} \hat{f}_a^\sigma. \quad (\text{A2})$$

This relation arises from the linearity of $\hat{f}_a = \sum_j t_{aj} c_j$, together with the notions that $\hat{f}_a^- \equiv \hat{f}_a$ and $\hat{f}_a^+ \equiv \hat{f}_a^\dagger$.

Let us start with the derivation of Eq. (A2). It is carried out by recognizing some basic quantum mechanics relations as follows. The first one is

$$e^A B e^{-A} = e^A B = \sum_{n=0}^{\infty} \frac{1}{n!} \mathcal{A}^n B, \quad (\text{A3})$$

where $\mathcal{A}B \equiv [A, B]$. We have therefore

$$e^{\beta\mu\hat{N}} \hat{f}_a^\sigma e^{-\beta\mu\hat{N}} = \sum_{n=0}^{\infty} \frac{(\beta\mu)^n}{n!} \hat{\mathcal{N}}^n \hat{f}_a^\sigma, \quad (\text{A4})$$

where $\hat{\mathcal{N}} \hat{f}_a^\sigma \equiv [\hat{N}, \hat{f}_a^\sigma]$.

We shall also use the quantum mechanics relations of

$$[AB, C] = A[B, C]_\pm \mp [A, C]_\pm B, \quad (\text{A5})$$

where $[\cdot, \cdot]_- \equiv [\cdot, \cdot]$ and $[\cdot, \cdot]_+ \equiv \{\cdot, \cdot\}$ denote the commutator and anticommutator, respectively. By noticing that $[c_k, c_j^\dagger]_\pm = \delta_{kj}$ and $[c_k, c_j]_\pm = 0$ for Fermions (+) or Bosons (−), we have $[\hat{N}, c_j] = \sum_k [c_k^\dagger c_k, c_j] = -c_j$ and $[\hat{N}, c_j^\dagger] = c_j^\dagger$. Thus, $[\hat{N}, \hat{f}_a] = -\hat{f}_a$ and $[\hat{N}, \hat{f}_a^\dagger] = \hat{f}_a^\dagger$; i.e., $\hat{\mathcal{N}} \hat{f}_a^\sigma = [\hat{N}, \hat{f}_a^\sigma] = \sigma \hat{f}_a^\sigma$. It immediately leads Eq. (A4) to Eq. (A2), which also implies

$$e^{\beta(h_B - \mu\hat{N})} \hat{f}_a^\sigma(t) e^{-\beta(h_B - \mu\hat{N})} = e^{\sigma\beta\mu} \hat{f}_a^\sigma(t - i\beta), \quad (\text{A6})$$

since $[h_B, \hat{N}] = 0$ and $e^{\beta h_B} \hat{f}_a^\sigma(t) e^{-\beta h_B} = \hat{f}_a^\sigma(t - i\beta)$.

The derivation of the detailed-balance relation, the second identity of Eq. (6), is now straightforward, by using the last identity of Eq. (A1), the trace-cyclic invariance, Eq. (2) and Eq. (A6). We have

$$\begin{aligned} C_{ba}^{(\sigma)}(-t) &= \text{tr}_B[\hat{f}_a^\sigma(t) \rho_B^{\text{eq}} \hat{f}_b^\sigma(0)] \\ &= \text{tr}_B[\hat{f}_a^\sigma(t) e^{-\beta(h_B - \mu\hat{N})} \hat{f}_b^\sigma(0) \rho_B^{\text{eq}} e^{\beta(h_B - \mu\hat{N})}] \\ &= \left\langle e^{\beta(h_B - \mu\hat{N})} \hat{f}_a^\sigma(t) e^{-\beta(h_B - \mu\hat{N})} \hat{f}_b^\sigma(0) \right\rangle_B \\ &= e^{\sigma\beta\mu} C_{ab}^{(\sigma)}(t - i\beta). \end{aligned} \quad (\text{A7})$$

The derivation of Eqs. (6), the symmetry and the detailed-balance relations in the time domain, is now completed. Their frequency-domain counterparts, Eqs. (7) with Eq. (8), are then followed immediately.

APPENDIX B: PATH INTEGRAL FORMALISM: DERIVATION

Let $U_T(t, t_0; \{f_a^\sigma(t)\})$ be the propagator of the total system and bath space, satisfying the Schrödinger equation of the total Hamiltonian of Eq. (1),

$$\partial_t U_T = -i[H(t) + \sum_{a,\sigma} W_a^\sigma f_a^\sigma(t)] U_T. \quad (\text{B1})$$

The total density operator at time t is given by

$$\rho_T(t) = U_T(t, t_0; \{f_a^\sigma(t)\}) \rho_T(t_0) U_T^\dagger(t, t_0; \{f_a^\sigma(t)\}). \quad (\text{B2})$$

Assuming it initially at time t_0 is [cf. Eq. (2)]

$$\rho_T(t_0) = \rho(t_0) \rho_B^{\text{eq}} \propto \rho(t_0) e^{-\beta(h_B - \mu \hat{N})}, \quad (\text{B3})$$

the reduced density matrix of primary interest is then

$$\begin{aligned} \rho(t) &\equiv \text{tr}_B[\rho_T(t)] \\ &= \text{tr}_B[U_T(t, t_0; \{f_a^\sigma(t)\}) \rho(t_0) e^{-\beta(h_B - \mu \hat{N})} \\ &\quad U_T^\dagger(t, t_0; \{f_a^\sigma(t)\}) \rho_B^{\text{eq}} e^{\beta(h_B - \mu \hat{N})}] \\ &= \left\langle U_T(t, t_0; \{e^{\bar{\sigma}\beta\mu} f_a^\sigma(t - i\beta)\}) \rho(t_0) \right. \\ &\quad \left. U_T^\dagger(t, t_0; \{f_a^\sigma(t)\}) \right\rangle_B \\ &\equiv \mathcal{U}(t, t_0) \rho(t_0). \end{aligned} \quad (\text{B4})$$

The third identity is obtained by using the trace cyclic invariance and the relations of Eq. (A6).

Consider now the path integral expression of the reduced Liouville-space propagator $\mathcal{U}(t, t_0)$, as defined in the last identity of Eq. (B4). Let $\{|\alpha\rangle\}$ be a basis set in the system subspace. We have

$$\begin{aligned} &U_T(\alpha, t; \alpha_0, t_0; \{f_a^\sigma(t)\}) \\ &= \int_{\alpha_0}^{\alpha} \mathcal{D}\alpha e^{iS[\alpha]} \exp_+ \left\{ -i \sum_{a,\sigma} \int_{t_0}^t d\tau W_a^\sigma[\alpha(\tau)] f_a^\sigma(\tau) \right\}, \\ &U_T^\dagger(\alpha', t; \alpha'_0, t_0; \{f_a^\sigma(t)\}) \\ &= \int_{\alpha'_0}^{\alpha'} \mathcal{D}\alpha' e^{-iS[\alpha']} \exp_- \left\{ i \sum_{\sigma, a'} \int_{t_0}^t d\tau W_a^\sigma[\alpha'(\tau)] f_{a'}^\sigma(\tau) \right\}. \end{aligned}$$

The action functional $S[\alpha]$ is related to the reduced system Hamiltonian $H(t)$ only. The system operators $\{W_a^\sigma\}$ have also been represented in path integral representation. On the other hand, the stochastic bath dynamics variables $\{f_a^\sigma\}$ remain in the operator form that involve in the time-ordered exponential functions.

Substituting the above path integral expressions for Eq. (B4) leads to Eq. (19), with the influence functional there being given by

$$\begin{aligned} \mathcal{F} &= \left\langle \exp_+ \left\{ -i \sum_{a,\sigma} \int_{t_0}^t d\tau W_a^\sigma[\alpha(\tau)] e^{\bar{\sigma}\beta\mu} f_a^\sigma(\tau - i\beta) \right\} \right. \\ &\quad \left. \times \exp_- \left\{ i \sum_{a',\sigma'} \int_{t_0}^t d\tau W_{a'}^{\bar{\sigma}}[\alpha'(\tau)] f_{a'}^\sigma(\tau) \right\} \right\rangle_B \\ &\equiv \exp \{-\Phi[\alpha, \alpha']\}. \end{aligned} \quad (\text{B5})$$

For $\{f_a^\sigma(t)\}$ satisfying Gaussian statistics, the bath ensemble average in Eq. (B5) can be evaluated exactly by using the second-order cumulant expansion method, as the higher order cumulants are all vanished. This property, together with Eq. (6), leads to the expression

$$\begin{aligned} &\Phi[\alpha, \alpha'] \\ &= \sum_{a,a'} \int_{t_0}^t d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 \left\{ W_a[\alpha(\tau_2)] W_{a'}^\dagger[\alpha(\tau_1)] C_{aa'}^{(+)}(\tau_2 - \tau_1) \right. \\ &\quad + W_a^\dagger[\alpha(\tau_2)] W_{a'}[\alpha(\tau_1)] C_{aa'}^{(-)}(\tau_2 - \tau_1) \\ &\quad + W_{a'}^\dagger[\alpha'(\tau_1)] W_a[\alpha'(\tau_2)] C_{aa'}^{(-)*}(\tau_2 - \tau_1) \\ &\quad \left. + W_{a'}[\alpha'(\tau_1)] W_a^\dagger[\alpha'(\tau_2)] C_{aa'}^{(+)*}(\tau_2 - \tau_1) \right\} \\ &\quad - \sum_{a,a'} \int_{t_0}^t d\tau_2 \int_{t_0}^{\tau_2} d\tau_1 \left\{ W_a[\alpha(\tau_2)] W_{a'}^\dagger[\alpha'(\tau_1)] C_{aa'}^{(-)*}(\tau_2 - \tau_1) \right. \\ &\quad \left. + W_a^\dagger[\alpha(\tau_2)] W_{a'}[\alpha'(\tau_1)] C_{aa'}^{(+)*}(\tau_2 - \tau_1) \right\} \\ &\equiv \int_{t_0}^t d\tau \mathcal{R}[\tau; \{\alpha\}]. \end{aligned} \quad (\text{B6})$$

for the influence functional exponent.

In detail, we have used the symmetry relation [the first identity of Eq. (6)] in the third and fourth terms, and the detailed-balance relation [the second identity of Eq. (6)] in the last two terms of the above expression. Some elementary algebra will lead to Eq. (B6), in terms of $\mathcal{R} \equiv \partial_t \Phi$, the expression,

$$\begin{aligned} \mathcal{R}[t; \{\alpha\}] &= \sum_{a,a'} \left(\{W_a[\alpha(t)] - W_a[\alpha'(t)]\} \right. \\ &\quad \times \{ \tilde{W}_{aa'}^{(+)}[t; \{\alpha\}] - \tilde{W}_{aa'}'^{(+)}[t; \{\alpha'\}] \} \\ &\quad + \{W_a^\dagger[\alpha(\tau)] - W_a^\dagger[\alpha'(\tau)]\} \\ &\quad \times \{ \tilde{W}_{aa'}^{(-)}[t; \{\alpha\}] - \tilde{W}_{aa'}'^{(-)}[t; \{\alpha'\}] \} \Big) \\ &= \sum_{aa',\sigma} \{W_a^\sigma[\alpha(t)] - W_a^\sigma[\alpha'(t)]\} \\ &\quad \times \{ \tilde{W}_{aa'}^{(\sigma)}[t; \{\alpha\}] - \tilde{W}_{aa'}'^{(\sigma)}[t; \{\alpha'\}] \}, \end{aligned} \quad (\text{B7})$$

with $\tilde{W}_{aa'}^{(\sigma)} \equiv \tilde{W}_a^{(\sigma)}$ and $\tilde{W}_{aa'}'^{(\sigma)} \equiv \tilde{W}_a'^{(\sigma)}$ being given by Eqs. (21). Using the definitions of Eqs. (20) and (22), the above expressions can be recast as Eqs. (23).

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